BOREAL ENVIRONMENT RESEARCH 12: 29–35
Helsinki 22 February 2007 ISSN 1239-6095

^{134}Cs and ^{137}Cs in lichen (*Cladonia stellaris*) in southern Finland

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Received 10 May 2006, accepted 23 Oct. 2006 (Editor in charge of this article: Veli-Matti Kerminen)

Puhakainen, M., Rahola, T., Heikkinen, T. & Illukka, E. 2007: ^{134}Cs and ^{137}Cs in lichen (*Cladonia stellaris*) in southern Finland. *Boreal Env. Res.* 12: 29–35.

The variation of the amounts of fallout radionuclides in reindeer lichen *Cladonia stellaris* (syn. *Cladonia alpestris*) and the underlying soil below the lichen was investigated in 1986–2004 at three locations in southern Finland. Samples from the lichen carpet were fractionated into three vertical layers and the distribution of radionuclides between the different fractions was investigated. The effective half-lives of ^{137}Cs in lichen were almost the same in all three layers and, as a whole, the effective half-life of lichen varied from 2.7 to 3.4 years.

Introduction

Lichens have been recognised as ideal indicators of airborne radionuclides in various ecosystems. They are long-living organisms composed of symbiotic fungi and algae, obtaining their nutrients mainly directly from the air (Mattsson 1975). Lichens effectively accumulate and retain most of the deposited fallout products and thus act as a reservoir of radioactivity.

They have no root system and are therefore unlikely to accumulate significant levels of ^{137}Cs from the substrate (Papastefanous *et al.* 1989). The uptake of radionuclides by lichens is enhanced by certain of their characteristics, such as the persistence of aerial parts slowing down their growth rates, long life spans and the high surface-to-mass ratio. The uptake of radioactive fallout products by lichens is rapid and nonselective allowing the immediate determination of short-lived isotopes (Ellis and Smith 1987).

Their scavenging properties make lichens cost-effective monitoring devices for aerosol reactive radionuclides. In modelling the transport of radioactive substances through lichens, it is assumed that the uptake occurs via the accumulation of radionuclides from the atmosphere. The removal occurs both via radioactive decay and, by first order, via biological elimination processes. The actual rate of growth does not affect the radionuclide inventory as long as the surface area increases linearly with time. Actual lichen growth alternates with comparatively dormant intervals (Smith and Ellis 1990).

Reindeer lichen (*Cladonia stellaris*) is also well known as the main source of intake of radionuclides, such as ^{137}Cs in reindeer. Lichen is the initial link in the food chain lichen–reindeer–man known to efficiently enrich radionuclides. From the point of view of radiation protection, it is especially important to determine the radionuclide content in the lichen carpet and the rate at which these radionuclides are eliminated from the carpet.

Since the beginning of the 1960s, the concentrations of ^{137}Cs in reindeer lichens have been

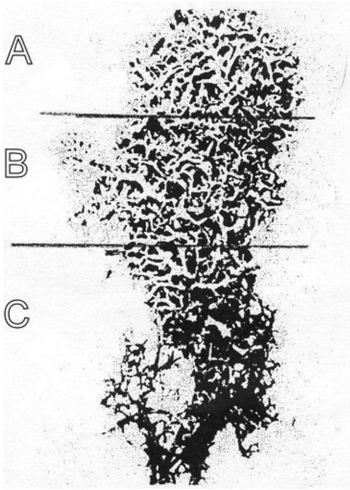

Fig. 1. Fractionation of lichen *Cladonia stellaris.*

investigated in Finland. In the 1960s, the highest mean concentration of ^{137}Cs in lichen in Finland was about 2400 Bq kg^{-1} dry weight (Tuominen and Jaakkola 1973), and in 1985 the ^{137}Cs concentration was approximately 200 Bq kg^{-1} (STUK 1987). After the Chernobyl accident, in 1986 and 1987 the ^{137}Cs concentration in the reindeer herding area in northern Finland varied from 200 to 2500 Bq kg^{-1} dry weight (K. Rissanen pers. comm.). An intensive investigation of the activity concentration of ^{137}Cs in lichen was carried out after the Chernobyl accident in the northern part of Finland because of the risks the radioactivity in lichen presented for the reindeer herding. The radioactive deposition deriving from the Chernobyl accident was very unevenly distributed in Finland. Fortunately the amounts of ^{137}Cs deposition in the vulnerable northern part of Finland were only about one-tenth of the corresponding amounts in southern Finland. No reindeer herding takes place at the highest fallout locations, but lichen has been transported from these areas to the northern parts for feeding reindeer. To obtain information about the content and distribution of radionuclides and the half-life of ^{137}Cs in lichens, samples of reindeer lichen were collected annually from 1986 to 2004 at three sites of high fallout deposition in southern and central Finland; Padasjoki, Loviisa and Olkiluoto. On 1 October 1987, the mean ^{137}Cs deposition in these municipalities was 77.7, 22.7 and 19.5 kBq m^{-2}, respectively (Arvela *et al*. 1990).

Material and methods

Samples of reindeer lichen (*Cladonia stellaris*) and the underlying soil (about 5 cm below lichen) were collected at three locations in southern and central Finland: in Padasjoki, the highest fallout area in Finland after the Chernobyl accident; in Loviisa, Hästholmen, near the nuclear power plant of Loviisa; and in Eurajoki, Olkiluoto, near the nuclear power plant of Olkiluoto. For practical reasons the sampling locations were chosen close to sites where also other studies were carried out. In Loviisa and Olkiluoto, the sampling sites were close to the sea in areas with rocks and stones, some barren trees and sparse undervegetation. Lichen grows on the rocks with very little soil below the lichen layer. The Padasjoki sampling site in central Finland is an area with many lakes of different sizes. Samples were collected from 1986 to 2004.

A standardised sampling technique was employed using a frame (25 × 25 cm) corresponding to an area of 0.0625 m^2. The samples were collected from the top of stones having little soil below the lichen. The mean dry weights of the soil below the lichen were 5.3 kg m^{-2} in Loviisa, 2.8 kg m^{-2} in Olkiluoto and 2.7 kg m^{-2} in Padasjoki. Parallel samples were collected when possible. Because reindeers prefer to eat only the fresh green top layer of the lichen carpet, it is important to examine the vertical distribution of the radioactivity in the lichen carpet and underlying material in order to estimate the transfer of radionuclides to reindeer. The lichen carpet was fractionated into four layers: fraction A (upper), fraction B (middle), and fraction C (lower) consisting mostly of the dead part of the lichen carpet (Fig. 1). The lichen samples were sprayed with water in the laboratory and all litter and foreign matter were removed from the lichen carpet with a pair of tweezers and combined into part D.

The samples were dried in an oven at 105 °C and homogenised, and the activities were measured gammaspectrometrically. The total uncertainty of the ^{137}Cs analysis (at the confidence level of 68%), including the calibration uncertainty of the measuring equipment and the statistical uncertainty of the measurement, was below 10%. The average dry weight of the cleaned lichen was 0.9 kg m^{-2} (Table 1).

In Padasjoki four soil samples from a nearby farming area were collected in 2004 to the depth of about 20 cm. The soil horizons were divided into layers, so that the first layer was 2-cm thick, the second layer was 3-cm thick and the other layers were 5-cm thick.

Results

In the highest fallout area, in 1987 the mean concentrations of ^{134}Cs and ^{137}Cs in cleaned lichen were 34 000 and 94 000 Bq kg^{-1} dry weight, respectively, whereas in 2004 the corresponding numbers were 2.5 and 1600 Bq kg^{-1} dry weight (Table 2). Besides the caesium isotopes, a few other relatively long-lived nuclides, including ^{106}Ru, ^{110m}Ag and ^{125}Sb, were detected in the samples taken between 1986 and 1989. Since 1987 the lichen samples were divided into vertical layers (Fig. 1). The highest concentration of ^{110m}Ag was detected in the topmost layer of lichen and that of ^{125}Sb in the lower layers (Table 3). The distribution of the ^{137}Cs concentrations in the layers of lichen in various years varied between the different locations (Fig. 2).

In the studied regions a small part of the ^{137}Cs concentration were the result of atmospheric nuclear weapons tests conducted in the 1950s and 1960s, but most of ^{137}Cs originated from the fallout of the Chernobyl accident in 1986. The highest activity in the total sample profiles found was 130 000 Bq m^{-2} in Padasjoki (Fig. 3). Most of the activity was detected in cleaned lichen (layers A, B and C) sampled during 2–3 years after the Chernobyl fallout. Later on, most of the activity was in the underlying soil and the D part (Figs. 3 and 4).

Table 1. Average dry weights of different layers of lichen (kg m^{-2}).

Part	Loviisa	Olkiluoto	Padasjoki	Average
A	0.282	0.258	0.258	0.263
B	0.257	0.279	0.288	0.277
C	0.282	0.343	0.369	0.339

Table 2. Concentrations of ^{134}Cs and ^{137}Cs (Bq kg^{-1} dry weight) in cleaned lichen.

Year	Padasjoki		Loviisa		Olkiluoto	
	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs
1986			10100	20800		
1987	34500	94400	8590	22100	3140	7780
1988	19400	69400	4000	14800	2450	8500
1989	11900	58600	4200	20300	1330	5960
1990	5000	34500	2100	14200	800	5090
1991	3970	36400	1330	12900	455	3880
1992			434	5440	177	2050
1993			311	5540	149	2410
1994	649	14000	171	4100	65	1360
1995	458	25900	73	2430	50	1540
1996	267	11200	31	1350	33	1440
1997			31	1290	20	1160
1998	58	4670	10	1060	2	943
1999	48	5600	2	1520		
2000	25	3330			5	710
2001	8	1610				
2002					0	269
2003	4.9	2090	0	813	0	318
2004	2.5	1560	0	449	0	308

Table 3. Concentrations of ^{106}Ru, ^{110m}Ag and ^{125}Sb (Bq kg^{-1} dry weight) in different layers of lichen.

Place/date	Layer of lichen	^{106}Ru	^{110m}Ag	^{125}Sb
Loviisa/29 July 1987	A	1200	150	330
	B	1700	89	330
	C	1800	82	450
Loviisa/30 July 1987	A	0	87	0
	B	0	0	0
	C	0	0	0
Loviisa/30 July 1987	A	790	94	0
	B	540	43	0
	C	260	0	140
Olkiluoto/20 May 1987	A	470	94	0
	B	740	0	230
	C	820	37	150
Olkiluoto/26 May 1989	A	0	0	0
	B	0	0	0
	C	200	0	85
Padasjoki/30 Oct. 1987	A	0	290	0
	B	0	280	1170
	C	0	180	1100
Padasjoki/30 Oct. 1987	A	0	230	0
	B	0	260	1400
	C	0	230	1300
Padasjoki/2 Oct. 1988	A	0	0	0
	B	0	0	0
	C	0	0	1100
Padasjoki/1 Oct. 1989	A	0	0	0
	B	0	0	300
	C	0	0	790

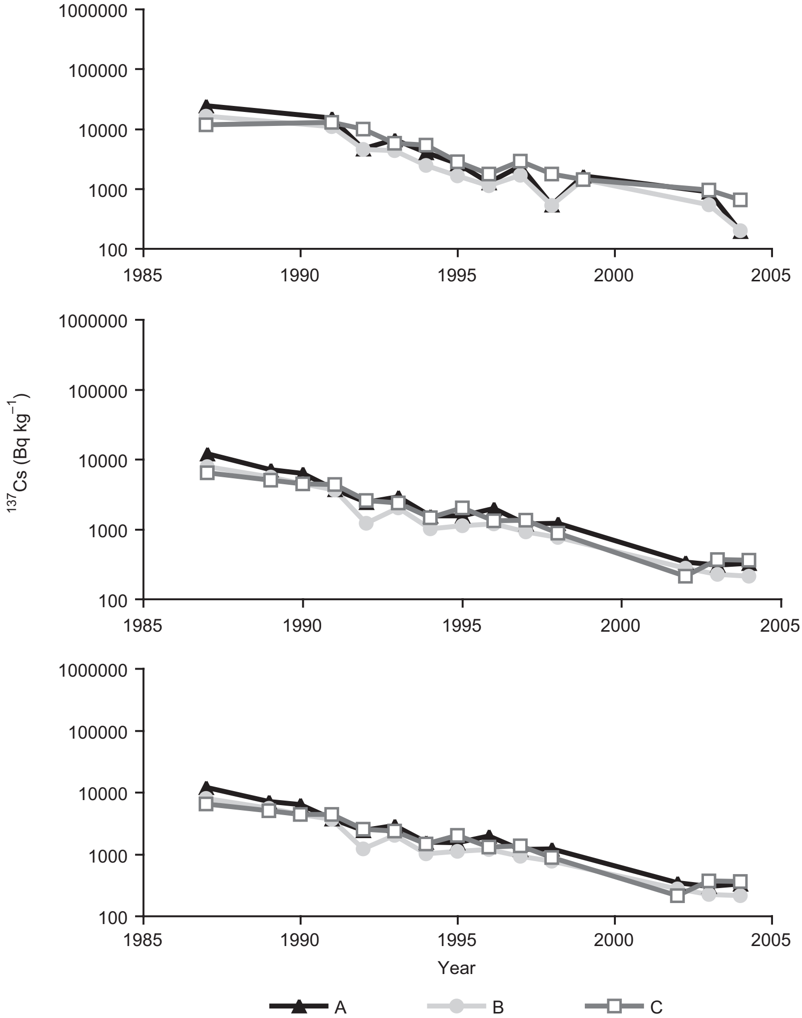

Fig. 2. Mean concentration of ^{137}Cs (Bq kg^{-1} dry weight) in different layers in lichen measured in Loviisa (top), Olkiluoto (middle) and Padasjoki (bottom).

In separate soil samples collected in Padasjoki in 2004, 48%–87% of the ^{137}Cs amount were found in the uppermost 5-cm layer and 85%–98% of it was in the uppermost 10-cm layer. Only 1.5%–14% of the ^{137}Cs concentration was in the lower layers.

Discussion

Lichen is among the most radiation-resistant living plants (Mattsson 1975). In the 1970s the maximum activity concentrations of all radionuclides, except ^{125}Sb, were found in the top layer of the lichen carpet. According to increasing depth penetration in the lichen carpet, the radionuclides may be arranged into the following sequence: ^{144}Ce ~ ^{7}Be << ^{95}Zr < ^{137}Cs < ^{106}Ru ~ ^{155}Eu << ^{125}Sb. The maximum values for the ^{125}Sb concentration were found in the B layer of the lichen carpet, not in the A layer, as was the case with ^{137}Cs and other radionuclides. This same was also observed in our studies conducted in 1987–1989 (Table 3). It seems that already a year after the deposition, ^{125}Sb was not found in the topmost part of lichen but it was transported to the lower parts and to the ground. According to Mattsson (1975), the reason for this was the low fallout rate combined with the very low solubility of ^{125}Sb that does not allow the upward movement with the liquid transport in the lichen plant, as in the case of ^{137}Cs (Mattsson 1975).

In Loviisa and Olkiluoto the highest amount of ^{137}Cs was in the topmost layer of lichen col-

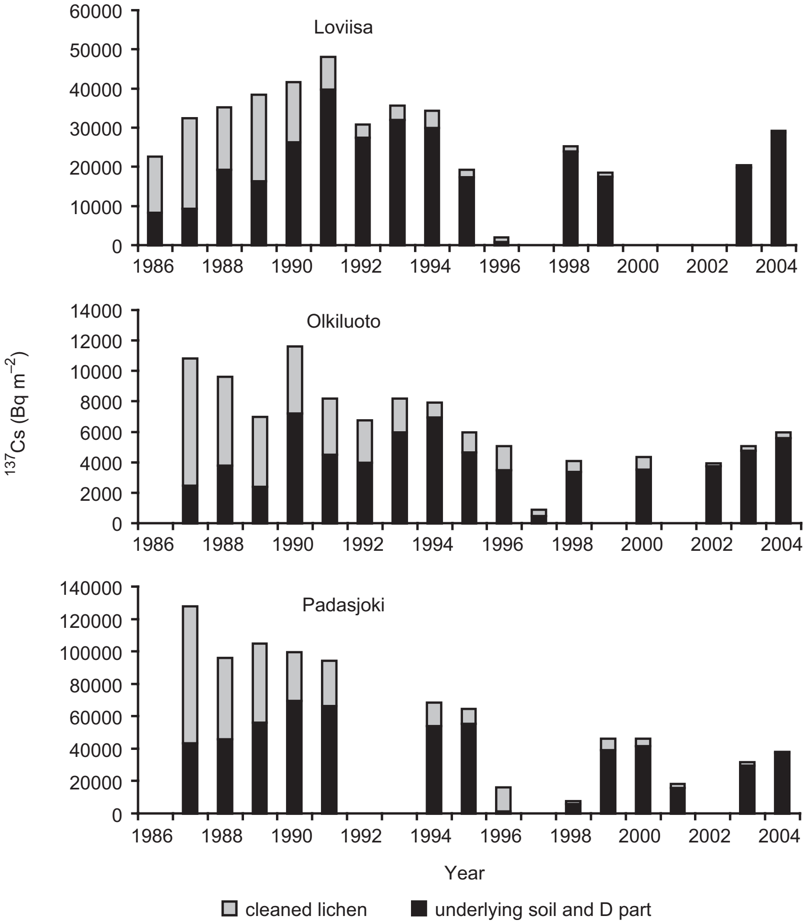

Fig. 3. Average amounts of ^{137}Cs (Bq m^{-2}) in the total profile and cleaned lichen.

lected during the four years after the Chernobyl fallout. In 1991, the amounts of ^{137}Cs were similar in all three layers. After 1991, the highest amounts of ^{137}Cs were detected in the lowest layer of lichen. Radiocaesium either moved into the lower layers of lichen or it was in the soil particles contaminating the lower lichen layers (Fig. 5). In Padasjoki, the highest fallout area of Finland, the variation was more erratic.

The effective half-life of ^{137}Cs in *Cladonia* species lichen, as presented in the literature in the 1980s and 1990s, varies from 1 to 10 years (Ellis and Smith 1987, Roos *et al*. 1987, Gaare 1990, Roos *et al*. 1991, Synnott *et al*. 2000). In a lake district in central Sweden, the effective half-life of nuclear bomb-produced ^{137}Cs in lichen was calculated to be about 8–10 years. A shorter half-life (7–8 years) was noticed in the upper fraction of lichen as compared with that in the whole lichen carpet (Roos *et al*. 1991). In our study the effective half-lives of ^{137}Cs from the Chernobyl fallout were almost the same in all three fractions and the half-lives in the total lichen varied from 2.7 to 3.4 years. No variation in half-lives between the different sampling locations was detected by us. The comparison of results obtained from samples collected before the Chernobyl accident with those collected after the accident is difficult. The share of ^{137}Cs that originated from the nuclear weapons test was in our samples so small that it can be considered negligible.

The highest uncertainty in our results is caused by the sampling and division of lichen into layers. If only a few years are taken into account when calculating the effective half-life, occasional variation in sampling can highly influence the results (*see* Fig. 2) (Sloof and Wolterbeek 1992). Many of the half-life calculations of lichen, as presented in the literature, were made using results that cover only a few years.

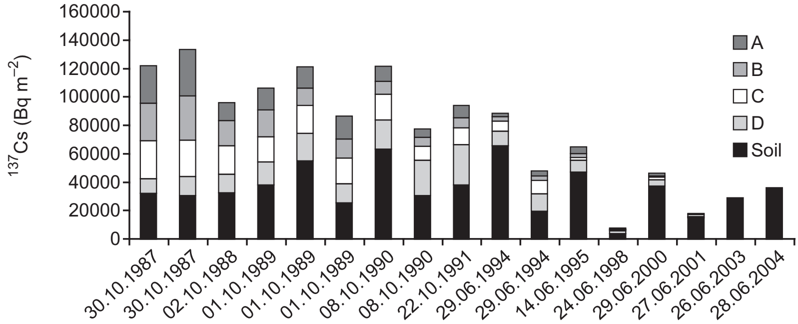

Fig. 4. Amounts of ^{137}Cs in different layers of lichen and soil beneath the lichen in Padasjoki. All parallel samples are presented separately.

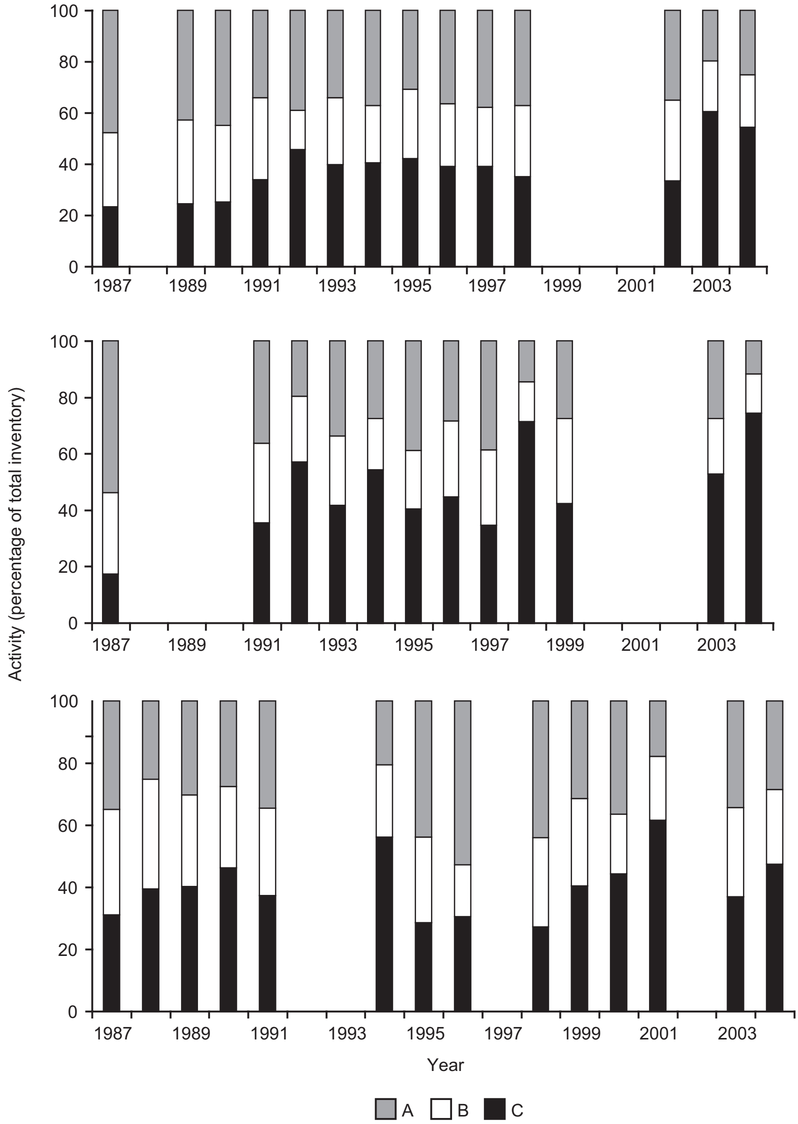

Fig. 5. Mean per cent of the total inventory of ^{137}Cs in different layers in lichen measured in Loviisa (top), Olkiluoto (middle) and Padasjoki (bottom).

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